Direct evidence for a dynamical ground state in the highly frustrated $Tb_2Sn_2O_7$ pyrochlore

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μSR experiments have been performed on powder sample of the "ordered spin ice" $Tb_2Sn_2O_7$ pyrochlore compound. At base temperature ($T = 35$ mK) the muon relaxation is found to be of dynamical nature which demonstrates that strong fluctuations persist below the ferromagnetic transition ($T_C = 0.87$ K). Hints of long range order appear as oscillations of the muon polarization when an external field is applied and also as a hysteretic behavior below T_C. We propose a dynamical and strongly correlated scenario where dynamics results from fluctuation of large spin clusters with the "ordered spin ice" structure.

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In the field of highly frustrated magnetism, rare-earth (RE) titanate pyrochlores $RE_2Ti_2O_7$ have attracted much attention in the last decade. As a result of large spin values of the RE atoms and usually small exchange coupling, spin anisotropy, dipolar and exchange interactions compete. The delicate balance of these energy scales, which varies with each particular pyrochlore compound, stabilizes original magnetic phases from spin ice (RE=Ho, Dy) to a unique collective paramagnetism in $Tb_2Ti_2O_7$ [1] still under debate. External constraints such as magnetic field or pressure can easily destabilize this balance and drive the system into complex phase diagrams [2, 3]. In the same trend of idea, special attention was recently devoted to the closely related stanate pyrochlore $RE_2Sn_2O_7$ compounds where lattice expansion as well as modification of the RE oxygen environment can also result in drastic changes of the balance of the interactions and eventually in novel exotic ground states [4]. Thus, while the titanate and stanate Tb pyrochlores exhibit similar antiferromagnetic correlations at high temperature, the Ti compound remains disordered and dynamical down to 70 mK whereas the stanate counterpart undergoes a "ferromagnetic" transition at 0.87 K. It was recently proposed from neutron experiments [5] that the stanate compound freezes in an original uniform ($\mathbf{q}$=0) spin ice structure where the four spins located at the vertices of each tetrahedron obey the "two in two out" ice rules [6]. The unexpected ferromagnetism would then result from the alignment of the spin vector sums on each tetrahedron. Interestingly, the Tb^{3+} frozen moment deduced independently from a nuclear Schottky anomaly in heat capacity measurements is nearly twice smaller than the one deduced from neutron scattering. Despite the static neutron picture, this suggested the existence of slow fluctuations, out of the neutron time window.

The coexistence of fluctuations, the fingerprint of frustration, and glassy behavior [7, 8, 9, 10] appears as a widespread and poorly understood feature of many frustrated systems. Even more surprising is the recently reported coexistence of spin dynamics and long range order, well below the transition temperature of some pyrochlore compounds (RE=Gd, Er) [11, 12, 13]. Using the μSR technique, we directly detect large spin fluctuations for the first time in the $Tb_2Sn_2O_7$ compound which we attribute to fluctuations of large spin clusters among the six-fold degenerate ground state of the ordered spin ice structure.

Positive muon μ^+ is a unique local probe to investigate directly spin fluctuations. With a large gyromagnetic ratio $\gamma_\mu = 2\pi \times 135.5$ MHz/T and a weak coupling to its magnetic surrounding, it is a very sensitive probe of magnetism. The accessible time window usually falls in between that of NMR and neutron experiments. As a noticeable example in the field of frustrated magnets, μSR gave the first direct evidence of a fluctuating ground state in the archetypal kagomé bilayers $SrCr_{9p}Ga_{12-9p}O_{19}$ [7]. In a μSR experiment, the asymmetry of the μ^+ decay between forward and backward positron detectors is recorded as a function of the muon life time in the sample. After subtraction of a background signal arising from muons which miss the sample, the asymmetry is directly proportional to the muon spin polarization $P(t)$. $P(t = 0)$ equals 1 since the muon beam is 100% spin polarized.

μSR measurements on powder samples of $Tb_2Sn_2O_7$ in zero and longitudinal applied field with respect to the muon initial polarization were performed at ISIS and PSI facilities. The samples were synthesized by standard solid state reaction and characterized by X-Ray diffraction at room temperature and SQUID temperature dependent susceptibility measurements.

The time dependence of the muon polarization has been recorded from room temperature down to 30 mK in a small longitudinal field $H_{LF} = 50$ G. It could be

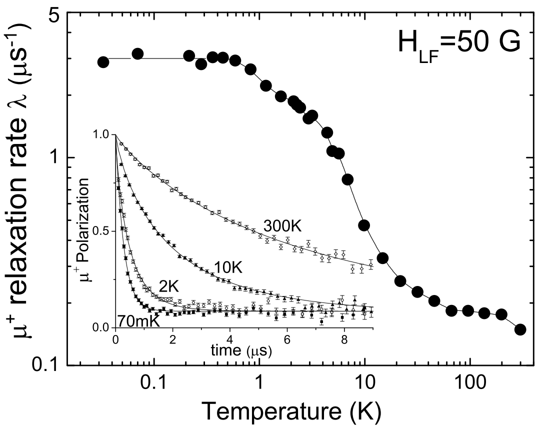

FIG. 1: Variation with temperature of the muon relaxation rate λ in a small 50 G longitudinal field on a $log-log$ scale. Inset: Muon polarization versus time at some selected temperatures, also in 50 G.

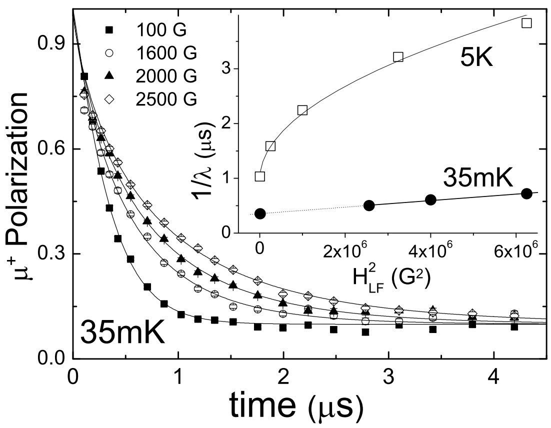

FIG. 2: Field dependence of the muon polarization relaxation at 35 mK for large applied fields. Inset: relaxation rates deduced from fits of the polarization with Eq. 1 (solid lines in main figure) plotted against H_{LF}^2 at 35 mK and for comparison at 5 K well above T_C. The lines are linear fits of $1/\lambda$ versus H_{LF}^2 at 35 mK and H_{LF} at 5 K.

fitted to a stretched exponential function

$$P(t) = e^{-(\lambda(T)t)^\alpha} + B \tag{1}$$

with a stretched exponent α close to 1 in the whole temperature range. The T independent $B \simeq 10\%$ term stands for the restored polarization of the muons which experience internal fields, typically of nuclear origin, much smaller than H_{LF} . The muon relaxation rate $\lambda(T)$ is presented in Fig. 1. For a single time relaxation, it is expected to be related to the electronic spin fluctuations rate ν by

$$\lambda = \frac{2\gamma_\mu^2 H_\mu^2 \nu}{\nu^2 + \gamma_\mu^2 H_{LF}^2} \tag{2}$$

where H_μ is the magnitude of the fluctuating field experienced by the muon. For zero or small external field, we thus get $\lambda \propto 1/\nu$. While the muon relaxation rate hardly depends on temperature at high temperatures, as a result of paramagnetic fluctuations, it steeply increases below 10 K and down to $\simeq 1$ K indicating a strong slowing down of spin fluctuations at the approach of the ferromagnetic transition at $T_C = 0.87$ K. Below T_C and down to the lowest temperature of the experiment $T = 30$ mK the muon relaxation rate saturates at a constant value. As depicted in the inset of Fig. 1, there is surprisingly no qualitative change in the shape of $P(t)$ above and below T_C. In particular, the usual signs of (i) a static ground state, namely the powder average long time tail $P(t \to \infty, T \to 0) = 1/3$, and (ii) long range order, namely oscillations of the polarization due to well defined internal field, are not observed.

In order to get more insight on the dynamical nature of the μ^+ relaxation, we studied the magnetic field dependence $P(H_{LF}, t)$ at base temperature. Were the relaxation at $T = 35$ mK due to a static local field H_μ at the muon site, or a distribution of width H_μ of such static fields in a more disordered scenario, H_μ would approximatively be given by λ/γ_μ. The full muon polarization $P(H_{LF}, t)$ should then be restored with $H_{LF}/H_\mu \gtrsim 5$. At 35 mK, $\lambda/\gamma_\mu \simeq 35$ G and as shown in Fig. 2 the relaxation is still strong under 2500 G applied field. Therefore we can safely conclude that the relaxation of the muon polarization is of dynamical nature. For a dynamical relaxation, much higher fields are needed than in the static case to suppress the T_1 processes. The field dependence is then given by Eq. 2. The field dependence of the muon polarization in $Tb_2Sn_2O_7$ appears to be rather complicated and we discuss it in more details in the following. However, if one restricts to high field and long time relaxation ($t \geqslant 0.1\mu$s) as in Fig. 2, $\lambda(H_{LF})$ nicely obeys Eq. 2 and we can extract $H_\mu \simeq 20$ mT and $\nu \simeq 0.2$ GHz in a straightforward manner. This fluctuation rate is below the accessible neutron diffraction time window (typically in the GHz to THz range). At higher temperatures, the $T_1 = 1/\lambda$ processes progressively evolve to an unexpected linear dependence with H_{LF} as shown in the inset of Fig. 2. Such a linear field dependence of $1/\lambda$ was formerly observed in $Tb_2Ti_2O_7$ [14, 15] and suggests similar dynamics in both compounds. In the case of $Tb_2Ti_2O_7$, this behavior was tentatively described by a power law decay of the spin time correlation function.

We now address the second feature of the low T relaxation, namely the absence of oscillations in zero external field. A basic scenario to reconcile long range order and dynamics is to assume that the spins fluctuate around the mean long range order in a magnon type picture. The absence of oscillations would then arise from very large and incoherent fluctuations so that the instantaneous distribution of the magnitudes of the internal field at the muon

site is wide enough to strongly damp the oscillations (in the extreme case of a fully disordered instantaneous field distribution one retrieves the well known dynamical Kubo-Toyabe polarization function which yields an exponential decay in the fast fluctuation limit [16]). Since the spin fluctuation rate ν is much lower than the neutron time window, neutron scattering experiments are sensitive to the instantaneous spin structure. The well defined magnetic Bragg peaks which yield a relatively large correlation length $l_C \simeq 18$ nm, together with the full value of the Tb moment detected at low T [5, 17], invalidate here such a basic scenario. We thus have to assume that muons experience a *well defined* internal field H_μ resulting from the well established ordered spin ice structure on the length scale l_C. In order to suppress oscillations of the muon relaxation, H_μ has to *fully* fluctuate in direction so that the mean internal field vector at the muon site cancels out. For a fluctuation rate $\nu \gtrsim \gamma_\mu H_\mu$, one then gets an exponential-like decay of $P(t)$ (see for instance a comprehensive calculation for this case in [15]). This is understandable here since the ordered spin ice state is six fold degenerate, the degrees of freedom being the choice of the two in and two out spins out of the four spins of one tetrahedron or equivalently of the direction of the resulting moment among one of the six (100) type directions. We thus propose that whole spin clusters, *i.e.* domains of typical size l_C where the tetrahedrons are all in the same configuration, fluctuate in between the six allowed configurations. Due to the high symmetry of the proposed ground state, the mean vector field at the muon site effectively vanishes. The correlation length l_C measured by neutron experiments [5] remains limited to 18 nm well below T_C. It is therefore consistent with the proposed cluster scenario. The small fluctuation rate ν we found also supports cluster fluctuations rather than single spin paramagnetic ones. In the proposed picture, the "ferromagnetic" transition corresponds to the freezing of the spin correlations on a large but finite length scale l_C which defines the extent of "long range" order. This order is dynamical in the sense that the ordered clusters undergo global rotations while the spin spin correlations are preserved.

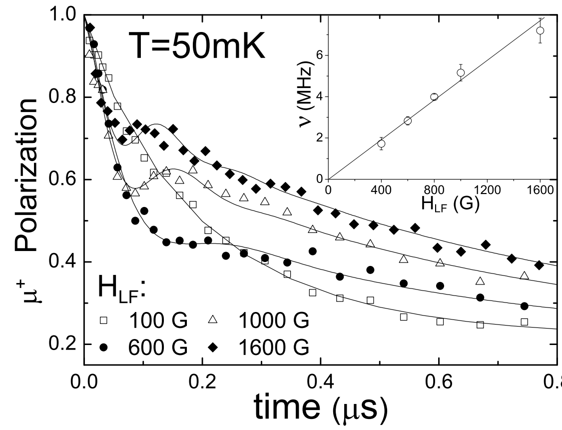

FIG. 3: Field dependence of the polarization relaxation for not too large fields. The solid lines are phenomenological fits to the sum of a damped cosine and a stretched exponential functions used to track the early time oscillation frequency ν plotted in the inset as a function of H_{LF}.

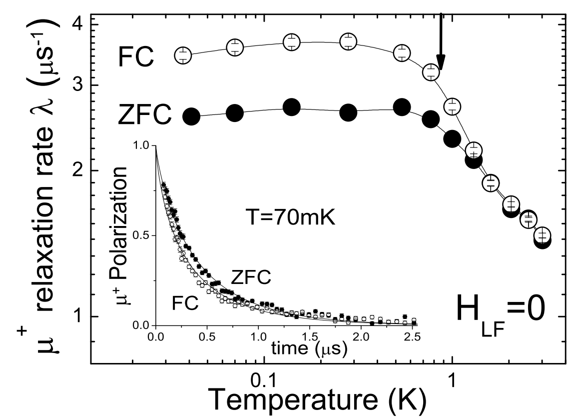

FIG. 4: Muon relaxation rate λ deduced from fits with Eq. 1 of the zero field polarization versus time data measured after cooling down the sample below 2 K in zero external field (ZFC) or with $H_{LF} = 800$ G (FC). The inset shows the raw zero field polarization at base temperature for the two cooling procedures.

In our μSR study, the "long range ordered" ground state of $Tb_2Sn_2O_7$ is only clearly evidenced in the detailed analysis of magnetic field effects on the μ^+ relaxation. As shown in Fig. 3, for intermediate values of the applied longitudinal fields H_{LF}, smaller than the ones used in Fig. 2, the relaxation is no more exponential but exhibits a strongly damped oscillation at early times. The frequency of this oscillation is plotted in the inset of Fig. 3. It scales linearly with the applied field according to $2\pi\nu/\gamma_\mu = A \times H_{LF}$ with $A \simeq 0.37$. A common situation where oscillations result from an applied longitudinal field appears in disordered magnets either above or below their transition temperature, when the field at the muon site H_μ is comparable to the applied field H_{LF}. The muon spin then precesses around the total field $\mathbf{H_{LF}} + \mathbf{H}_\mu$ and due to the disordered nature of H_μ, either dynamical or static, the resulting polarization shows a highly damped oscillation at a frequency $\gamma_\mu H_{LF}/2\pi$[16, 18]. The small value of A rules out such an explanation here and the oscillations are more likely intrinsic, *i.e.* they arise from "long range order" in agreement with the proposed cluster scenario. A should then rather be associated with a magnetic susceptibility resulting from progressive spin canting along the applied field. The effect of the external field is to break the high symmetry of the ground state. The mean vector field at the muon site is no more zero and oscillations are observed.

After applying increasing longitudinal fields at base temperature as described above, we surprisingly did not recover the same polarization decay in zero field. Namely, the relaxation rate was slightly higher after the field experiment than before, although the polarization versus time curves are qualitatively similar (see inset in Fig. 4). Relatively strong applied field ($H_{LF} \gtrsim 800$ Oe) are likely to drive the system in a different spin configuration resulting in a higher field $H_\mu + \Delta H_\mu$ at the muon site. Considering the small magnitude of ΔH_μ (a fit with Eq. 1 gives $\Delta H_\mu / H_\mu \simeq 15\%$ at 35 mK) one can assume that only minor modifications of the overall ordered spin ice structure subsist after application and removal of the magnetic field. We checked that the effect of an external magnetic field as presented in Fig. 2 and 3 hardly depends on the initial zero field state. Thus, despite a slight change in the spin configurations, the ground state remains dynamical. Such an hysteresis effect is a signature of frozen spin correlations as we expect below T_C. Field history dependence have for instance been reported in the spin ice $Ho_2Ti_2O_7$ compound in neutron [20] and μSR [21] experiments. We further investigated this effect by measuring the muon relaxation rate as a function of temperature from 70 mK up to 3 K in zero external field after (i) cooling down the sample from 3 K in zero external field and (ii) cooling down the sample with H_{LF}=800 G and then removing the external field at base temperature. The results are plotted in Fig. 4. Interestingly the hysteresis starts at a somewhat higher temperature $T \simeq 1.3$ K than the actual transition temperature defined by the peak in the heat capacity measurements at $T_C = 0.87$ K. This perfectly agrees with neutron measurements where the magnitude of the frozen Tb moment was shown to vanish only above 1.3 K. These results point at a rather broad transition regime.

To summarize, we have evidenced that a dynamical regime survives in $Tb_2Sn_2O_7$ far below its ferromagnetic transition. This compound therefore joins the recently exhibited class of pyrochlores where fluctuations and long range order are simultaneously observed, namely $Gd_2Ti_2O_7$, $Gd_2Sn_2O_7$ and $Er_2Ti_2O_7$. Our results for $Tb_2Sn_2O_7$ are however markedly different from the $Gd_2Ti_2O_7$ case, where fluctuations were observed in addition to oscillations of the muon relaxation. Here the exponential decay of the muon polarization cannot be ascribed to fluctuations around an overall long range order but results from complete fluctuation of the local field. To get a consistent picture with neutron results [3], we proposed that whole clusters of well ordered spins fluctuate in between the six degenerate configurations allowed in the "ordered spin ice" structure. Application of an external longitudinal field breaks the symmetry of the configuration sub-space and oscillations can be detected. Frozen correlations are also evidenced by a field history dependence of the zero field muon relaxation below $\simeq T_C$.

The large number of spins involved in the coherent rotation of a cluster demand a unique configuration space with vanishingly small energy barriers in between the six fold degenerate configurations of the ground state in order to allow for low energy collective excitations. This is rather counterintuitive with respect to the Ising type anisotropy which stabilizes the local spin ice order. Contrary to spin ice pyrochlores, the "ordered spin ice" structure may not be dominantly driven by single ion anisotropy and the latter may not be too strong in $Tb_2Sn_2O_7$. It is also noticeable that, despite the ferromagnetic transition of $Tb_2Sn_2O_7$, the muon relaxation is strikingly similar to the well studied parent compound $Tb_2Ti_2O_7$. One may assume that the dynamics observed here is not different in nature from the collective excitations of the disordered $Tb_2Ti_2O_7$ magnet [1, 14]. Understanding the exotic dynamical ground state of $Tb_2Sn_2O_7$ where correlations are well defined may in turn bring new insight to the puzzling and more complex case of the Ti counterpart.

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